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Evaluation of the wavelengths of pulsed laser induced fluorescence spectrum by simultaneous recording of standard emission lines

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Abstract : A simple experimental technique for calibration of unknown wavelengths of pulsed laser induced fluorescence (PLIF) spectrum has been demonstrated by simultaneous recording of PLIF spectrum and standard emission spectrum from hollow cathode lamp (HCL). Both PLIF and HCL signals are detected by the same photomultiplier tube and separated by employing gated and phase sensitive detection techniques

Keywords : Calibration, laser induced fluorescence, gated and phase sensitive detection.

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The monochromator, extensively used in laser spectroscopic research needs to be calibrated for evaluating the wavelength of unknown spectral lines. Most of the monochromators utilise the mechanical counter system which approximately determines the wavelength, and in some calibration is provided by using optical encoders. The wavelength reproducibility of the counter reading of the calibrated monochromator with some standard light source is usually not very good, especially for low dispersion-high speed monochromator usually used in fluorescence studies. In order to have a reliable calibration, one may need the simultaneous recording of standard spectrum along with the unknown spectrum on the same strip chart so that one can find out the wavelength of unknown spectral lines precisely.

A simple experimental technique for calibration of unknown wavelengths of laser induced fluorescence (LIF) spectrum has been demonstrated by simultaneous recording of LIF spectrum and standard emission spectrum from hollow cathode discharge lamp (HCDL). The chopped light from copper-neon hollow cathode lamp and the pulsed-laser induced fluorescence (e.g. Ba atoms) are allowed to fall on the monochromator slit, both these signals are then detected by the same photomultiplier tube (PMT) placed at the exit slit of the monochromator. The PMT signals are then separated by simultaneously employing gated and phase sensitive detection techniques and recording both LIF and HCDL signals on a multipen strip chart recorder. The accurately known wavelengths of the emission lines of Cu and Ne thus provides the chart dispersion, which is then used to evaluate the wavelength of unknown laser induced fluorescence lines of barium. The calibration of dye laser wavelengths using optogalvanic effect in Ne HCDL has been carried out by many workers (e.g. Camus *et al* 1989 and Syage *et al* 1985).

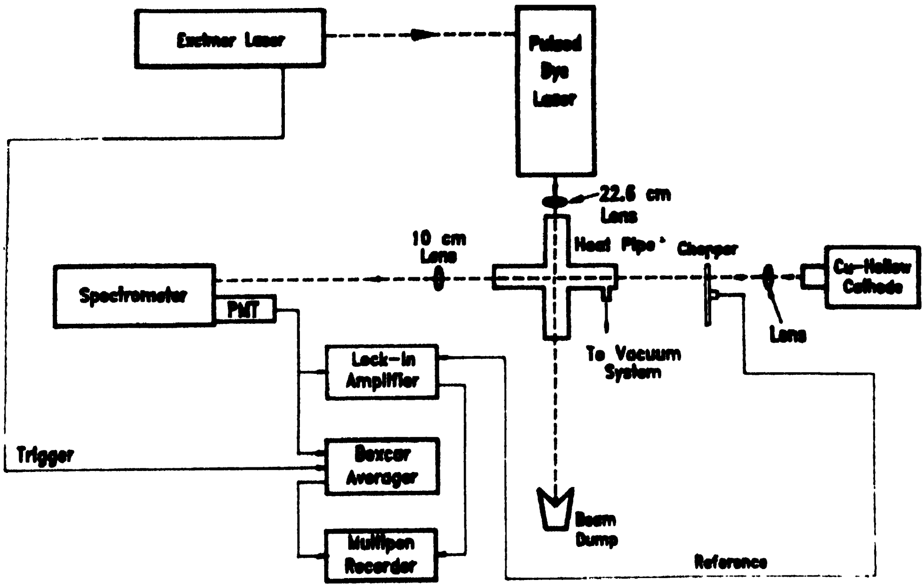

Figure 1. Schematic experimental layout showing simultaneous recording of pulsed laser induced fluorescence spectrum and calibration spectrum from a hollow cathode discharge lamp.

Multiphoton ionisation of Ba atom in heat-pipe oven using high power excimer laser (Lamda Physik, EMG 302 MSC) pumped pulsed dye laser (Lamda Physik, FL 3002) is

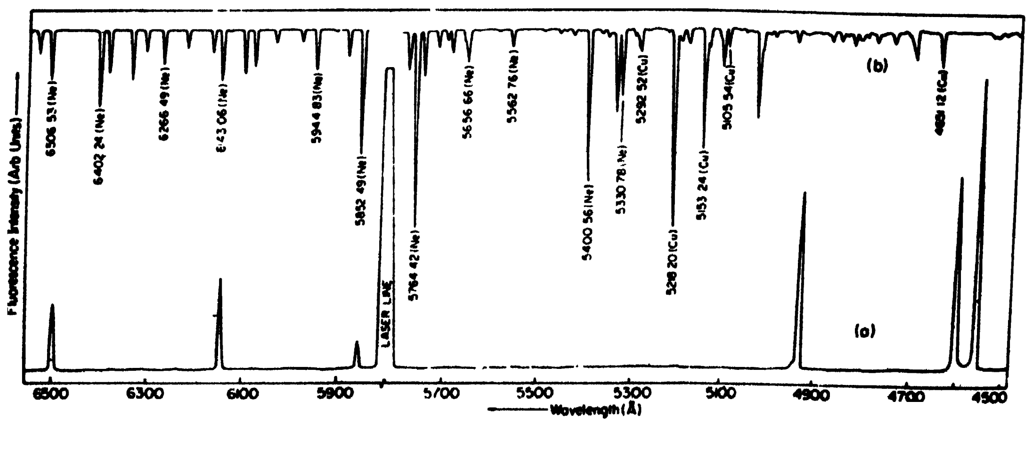

Figure 2. (a) Monochromator scan for a laser excitation wavelength of 5817.3 A° which is in resonance with two-photon transition $6s^{2\,1}$ $6p^{2\,1}S_0$; (b) Cu/Ne hollow cathode emission spectrum for wavelength calibration.

studied by monitoring fluorescence from barium atom and ions (for details see Nakhate *et al* 1991). The schematics of the experimental setup is shown in Figure 1. The laser induced fluorescence light of Ba is focused on the entrance slit (150 micron) of 0.5 meter monochromator (M/s Pacific Ins.) along with the light from Cu/Ne hollow cathode lamp as shown in Figure 1. The light from Cu/Ne hollow cathode lamp is mechanically chopped at frequency about 210 Hz (period = 4.7 msec). The pulsed dye laser is operated at repetition rate of 9 Hz. The output of the photomultiplier tube (PMT) of the monochromator, detecting both LIF and light from HCDL, is simultaneously fed to boxcar averager (EG & G, Model 162) and lock-in amplifier (Ithaco Dynatrac, Model 391A). The boxcar averager

Table 1. Evaluated wavelengths of pulsed laser induced fluorescence lines of Ba, using known emission lines of Cu/Ne (HCDL) for calibration. Laser wavelength fixed at two-photon transition in Ba I at λ = 5817.30 A° ($6s^{2}\,{}^{1}S_{0} \to 6p^{2}\,{}^{1}S_{0}$) and monochromator scanned.

Evaluated wavelength λ_L (in A°)	Listed* wavelength λ_A (in A°)	($\lambda_E - \lambda_A$) (in A°)	Assignment*			Origin
4554.19	4554.03	0.16	0.0 ($6s\,{}^{2}S_{1/2}$)	—	21952 ($6p\,{}^{2}P_{3/2}$)	Ba II
4599.56	4599.75	-0.19	12637 ($6s6p\,{}^{3}P_{1}$)	—	34371 ($6p^{2}\,{}^{1}S_{0}$)	Ba I
4934.30	4934.09	0.21	0.0 ($6s\,{}^{2}S_{1/2}$)	—	20262 ($6p\,{}^{2}P_{1/2}$)	Ba II
5853.84	5853.68	0.16	4874 ($5d\,{}^{2}D_{3/2}$)	—	21952 ($6p\,{}^{2}P_{3/2}$)	Ba II
6142.04	6141.72	0.32	5675 ($5d\,{}^{2}D_{5/2}$)	—	21952 ($6p\,{}^{2}P_{3/2}$)	Ba II
6496.72	6496.90	-0.18	4874 ($5d\,{}^{2}D_{3/2}$)	—	20262 ($6p\,{}^{2}P_{1/2}$)	Ba II

* From Meggers *et al* (1975). In column 4 the energy level values are in units of cm^{-1}.

is triggered by synchronous pulse from excimer laser power supply unit and the chopper output is given as a reference for lock-in amplifier. The gate width of the boxcar averager is kept a few nanosecond. In this way we could separate the signals because of pulsed laser induced barium fluorescence and also of Cu/Ne hollow cathode light. The outputs from the boxcar averager and lock-in amplifier were recorded on a multipen strip chart recorder. One such trace obtained at laser excitation wavelength of 5817.30 A°, which is in two-photon resonant from ground state $6s^{2}\,{}^{1}S_{0}$ to $6p^{2}\,{}^{1}S_{0}$ state of Ba atom, is shown in Figure 2. Since the boxcar gate width was few nanosecond and the chopped HCDL light source has a period of few milliseconds which is almost continuous in comparison to nanosecond gate width of boxcar amplifier, the HCDL light was loading the boxcar amplifier. In order to avoid this problem the HCDL light intensity was reduced to a minimum by using neutral density filters. We also found that the strong line due to LIF of Ba slightly loading the lock-in amplifier and vice versa, but this was not obscuring the simultaneous recording of both the signals (LIF and Cu/Ne lines) when they were either in the vicinity of each other or even overlapping. Using the well known emission lines of Cu and Ne (Reader and Corliss 1980) emitted by Cu/Ne hollow cathode lamp we obtained the chart dispersion

(A°/mm) and consequently evaluated the wavelengths of barium fluorescence lines which could be assigned to either atomic (Ba I) or ionic spectrum (Ba II). The evaluated wavelengths of LIF lines of Ba is given in Table 1 along with the listed wavelengths of Ba I and Ba II taken from Meggers *et al* (1975). The error in evaluation of the wavelength is of the order of 0.2 A°, which is quite accurate considering the fact that a low dispersion (17.5 A°/mm) monochromator was used. The fluorescence lines of Ba^+ ion originating from $6p\,^2P_{3/2,\,1/2}$ state are due to four-photon ionisation of Ba at this laser wavelength of 5817.30

To the best of our knowledge this is the first demonstration of an experimental technique for simultaneous recording of standard emission spectrum for evaluating the wavelengths of unknown lines of pulsed laser induced fluorescence spectrum. The technique is simple and gives reliable results.

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